Review of Plutonium Attribute Measurement Technologies Presentation on Neutron Measurements: Pu Mass & Absence of Oxide

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The presentation discusses the passive neutron measurements as applied to the attributes of plutonium Mass and the Absence of Oxide. The neutron multiplicity measurement results in the ²⁴⁰Pu-effective mass (m_{240}). The test for oxide is based upon α , which is the ratio of (α ,n) reactions to spontaneous fission. The neutron multiplicity system also provided outputs for the symmetry analysis, which is described in another presentation.

Any measurement of plutonium mass, whether it is an active or a passive technique (including calorimetry), requires isotopic data to extract the total plutonium mass. The total mass of plutonium is determined by combining the results of the isotopic data (PU600) and the m_{240} . Since the PU600 only relays the 240 Pu/ 239 Pu ratio, assumptions about the other plutonium isotopes may result in a bias.

Neutron coincidence counting has been used by the IAEA for many years. This is basically the process of relating m_{240} and doubles rate. This method, however, requires representative standards to calibrate the system. Problems arise when you assay materials that differ from your representative samples (i.e. the multiplication and/or α are different). The benefit of multiplicity counting is that it does not need representative calibration standards. A detector can be characterized using non-sensitive materials.

In order to perform neutron multiplicity measurements, three things are needed. First an optimized detector is needed with a high efficiency (typically 40-55%), a low die-away time ($< 30\mu s$), and small dead time. These parameters tie directly to the accuracy of the measurement and the counting time required to reach a given precision. Electronics are needed to acquire and analyze the signals from the counter. A multiplicity shift register does the time series analysis of the neutron pulse stream. Finally a mathematical model (with assumptions) is used to interpret the measured value. For the multiplicity shift register, we use the reactor "point model" to relate the singles, doubles and triples rate to 240 Pu –effective, α , and Multiplication (M).

The neutron multiplicity counter can be characterized using a well-known ²⁵²Cf source instead of having to produce costly representative standards. A detector parameters can be determined based upon the sources strength and nuclear parameters. The ²⁵²Cf source also provides a measurement control capability for determining that the system is function properly. Typically this is done by decay correcting the source rate and comparing to the new measurement. In this case we did not have an internal clock or

storage capabilities. We used the Doubles to Singles (D/S) ratio, which is fixed and independent of source strength, for determining that the system was operating properly. If the D/S ratio had changed, then this would imply that something was wrong with the system. One should point out that the efficiency that is determined using ²⁵²Cf is slightly different from that of plutonium because of the difference in fission energy spectra.

To make high quality measurements of plutonium, an optimized system would look something like the Large Neutron Multiplicity Counter (efficiency \approx 42%). The IAEA and domestic safeguards at LLNL have used this system for verification measurements. This system has already been used to measure weapons components at RFETS in ALR-8 containers. This proof-of-principle exercise showed that assay of components was possible. The comparison of assay to reference values for the 18 measurements, shows a scatter of \sim 6% for 30 minute runs. Some of the scatter is because of container effects. If the efficiency of the system were lower, then the count time would be longer to achieve the same precision.

The counter used in the FMTT Demo was a commercially available shipper/receiver counter. It was adequate for the purposes of the demonstration. An optimized version, however, would take less measurement time, incorporate the gamma system, give better precision and accuracy, and be more robust over sample to sample variations. This system is a single row counter with eight banks of four ³He tubes in a polyethylene matrix. The inner cavity and outer wall are lined with cadmium.

Up to this point, we have mainly focused on the measurement of mass. The NMC also ties into the "Absence of Oxide." The presence of oxygen in the system causes the production of (α,n) neutrons. Pure metal samples yield zero (α,n) neutrons. A "non-zero α is a potential indicator of oxide, but the presence of materials other that plutonium also affect the ratio. A non-zero α can result from the presence of other low-Z elements such as fluorine, magnesium, aluminum, or beryllium. Pure, freshly separated, low-burn-up plutonium oxide has an α typically greater than 0.5. This value assumes only ²³⁹Pu and ²⁴⁰Pu are present in the material and the isotopic ratio is less than 0.10.

Although calorimetry is considered the "gold standard," the neutron multiplicity counting technique is accurate and robust. Large insulating containers and the time needed to perform the measurement would hamper the practicality of using calorimetry in this application. This is a proven technology in use for international inspections and can be authenticated with non-sensitive materials. The system is capable of distinguishing plutonium for isotopic neutron sources such as ²⁵²Cf or AmLi. It does, however, require accurate isotopic data to make a total mass determination.